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Final Report

Air Force Office of Scientific Research
Contract No. F49620-91-C-0091

Femtosecond Photonics: Fundamental Phenomena and Device Behavior

Principle Investigators:

Professor James G. Fujimoto
Professor Hermann A. Haus
Professor Erich P. Ippen

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FINAL REPORT

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Introduction

This report is comprised of brief summaries of work accomplished on a number of topics related to femtosecond photonics. It describes significant progress in the development of new sources of ultrashort pulses, important advances in the understanding of ultrafast semiconductors and fiber optic device behavior, and new insights into fundamental, underlying phenomena in photonic materials. The summaries emphasize the most important and most recent results. A complete list of the 59 publications acknowledging this AFOSR support over the past three years is appended.

Fiber Ring Lasers

Several new fiber ring laser systems have been developed over the past few years by our group. The simplicity of design of these systems allows for modifications that lead to outstanding performance characteristics.

The lasers consist of erbium doped fiber and undoped fiber forming a ring. The net dispersion (group velocity as a function of frequency) can be adjusted through proper choice of the two fiber segments. Polarizers and quarter wave plates control the polarization of the mode circulating in the ring. An isolator assures propagation in one direction around the ring. By proper adjustment of the polarizers, the self-phase modulation of the two polarizations of the mode circulating in the ring is transformed into amplitude modulation such as to spontaneously form pulses in the ring by injection locking of the modes of the ring resonator (modelocking). Pulses are formed when higher intensities experience less loss than lower intensities. We call this mechanism additive pulse modelocking (APM), and this acronym is now accepted in the literature as the descriptor of this form of modelocking[1]. The ring delivers pulses of the order of 400 fs duration at the round-trip rate of 40 MHz. We have studied the wavelength tuning and filtering characteristics of this laser[2] and developed optimization criteria[3]. Last year, with proper adjustment of the fiber dispersion, we were able to produce pulses as short as 69 fs[4]. The system has

been copied by groups at Bell Laboratories and MIT Lincoln Laboratory for their use. We are currently in the process of working with Clark MXR to commercialize these systems.

The simplicity of these systems permits modifications for the realization of different operating characteristics. For many purposes it is desirable to generate pulses at much higher rates than 40 MHz. This can be accomplished with a modulator in the ring, yet not without further precautions. Indeed, a modulator by itself produces pulse trains of very poor pulse-to-pulse stability, because the gain relaxation time is of the order of a millisecond, and there is no mechanism to regulate the energy from pulse to pulse. Last year, we reported operation of a harmonically modelocked system operating at 1 GHz, using a modified APM mechanism, used to stabilize the energy from pulse to pulse[5]. Instead of providing decreasing loss for increasing intensity, the APM mechanism was reversed by a change of the polarization controllers to limit peak intensities. In this way, we obtained stable pulse-streams at 1 GHz, the highest frequency accessible with our modulator. MIT Lincoln Laboratory has copied our system and, with their 20 GHz modulator, produced stable pulse streams at this much higher repetition rate.

The modelocked ring laser described above delivers pulses of width according to the formula for active modelocking, dictated by the magnitude of the modulator drive which is limited. In the 1 GHz system just described, the pulse widths are 15 ps. In an attempt to generate shorter pulses at a high repetition rate, Chris Doerr, a graduate student, discovered asynchronous modelocking, a new principle of modelocking fiber ring lasers[6]. A phase modulator is used instead of the amplitude modulator described before. The modelocking drive of 1 GHz is detuned by about 15 kHz from the frequency synchronous with a harmonic of the round-trip frequency. The pulses are now 1 ps long and travel around the loop at their own rate. The modelocking drive serves the purpose of starting the system and of arranging the pulses equidistantly around the loop by refreshing the pulse stream every time it becomes synchronous with the modelocking drive. Because phase modulation is used, instead of amplitude modulation, the pulses do not experience modulator induced loss when they drift out of alignment with the modelocking drive. The APM action in the loop is responsible for the shortening of the pulse to 1 ps.

This is an entirely new principle of modelocking made possible by the pulse-forming capabilities of the fiber loop. The APM mechanism of the fiber loop and soliton shaping effects provide the short pulse widths. The limiting effects of their actions provide the stability for the pulse-to-pulse energy. If any conventional modelocked system were detuned in the same way, modelocked operation would simply cease. We believe that this asynchronous modelocking principle is applicable to all systems with their own passive pulse-shaping mechanism (e.g., Kerr Lens Modelocking) if harmonic modelocking with short pulse widths is sought.

Fiber Storage Ring

In a theoretical paper published in 1992[7], we proposed that a fiber loop with gain, filter, and an amplitude modulator could maintain a stream of pulses separated by empty

intervals (ones and zeros), in spite of the spontaneous emission noise associated with gain. In this way, a fiber ring could act as a storage ring of a bit-stream of optical pulses. This year, we demonstrated the storage principle with an experimental fiber ring containing 66 bits at a 1 GHz rate[8].

The system consists of two rings; one serves as the pulse pattern generator, the other as the storage ring. The pulse pattern generator is simply a ring laser with a modelocking element which, starting from noise, reaches a steady state with pulses in some of the time slots and "zeros" in other slots. Through a $LiNbO_3$ switch this pattern is injected into the storage ring. Approximately 2 pJ of injected energy is needed to establish a "one" in the storage ring. The "ones" are then renormalized to energies of about 50 pJ in the storage ring, and the "zeros" are further suppressed. The storage was on a time scale of hours (as long as the gain was maintained). We have addressed the stability and timing issue theoretically[9], and MIT Lincoln Laboratory, interested in high bit-rate optical storage, has built a storage ring according to the same design and stored an 18 Gb/s bit-stream. No physical principle prevents the upscaling of this storage to bit-rates of 100 Gb/s, if 100 GHz modulators become available.

Additive Pulse Modelocking

The principle behind additive pulse modelocking (APM) involves splitting a pulse into two pulses which accumulate differential phase profiles. One pulse is sent through a nonlinear medium and accumulates an intensity-dependent phase profile. The pulses are then interferometrically superimposed so that they add constructively at the peak and destructively in the wings. This produces a shorter pulse. This artificial fast saturable absorber action can be achieved in a single-cavity (via a Mach-Zehnder interferometer or interference of two polarizations) or a coupled-cavity arrangement. In our $NaCl:OH^-$ laser system, we use a coupled-cavity scheme with a gain medium in the main cavity and a nonlinear fiber in the auxiliary cavity.

Recently, we observed an "overdriven" operation of our $NaCl:OH^-$ APM laser[10] in which the laser operates very stably, producing shortpulses with very high, nonlinear phase shifts in the auxiliary fiber. This mode of operation is difficult to analyze with our conventional APM theory which assumes small nonlinear phase shifts in the fiber. In order to understand this mode of operation, we developed a new analytical theory which looks at this overdriven case[11]. In this theory, we propose that filtering provided by bulk optical elements in the laser allows the laser to operate with these large nonlinear phase shifts. Our theory takes the usual Master Equation of APM and treats the operation of the auxiliary cavity with a new operator. Furthermore, we use Gaussian-shaped pulses as an approximate solution in order to simplify the mathematics. Using this theory along with numerical simulations, we obtain results which are in good agreement with experimental results.

Another feature of this overdriven system is a hysteretic behavior we observed which was uncharacteristic of conventional APM systems. We observed a direction-dependence

of the laser operation depending on whether the auxiliary cavity length was lengthened or shortened. While the cavity length is lengthened, ordinary APM behavior occurs. Pulsing is intermittent, recovering with each wavelength of detuning. However, when the cavity is shortened, pulsing begins and persists over several wavelengths. In addition, the output pulses experience a spectral red-shift as their intensity increases. We have proposed an explanation for this behavior which is based upon the saturation of the artificial fast saturable absorber action[12] and the nonlinear gain curve for a self-limited additive pulse modelocked laser[13].

A Broadband-Tunable Femtosecond Source for 1.55 μm Diagnostics

Modern lightwave communication systems use carrier wavelengths around 1.55 μm because optical fibers have minimum losses and can be tailored to have negligible dispersion in this spectral region. Since all-optical systems are desirable, other communication system components need to operate around this wavelength as well. These include semiconductor laser diodes, detectors, optical amplifiers, and modulators. The characteristics of these components depend strongly on the material they are made of, the structure of the device, and the wavelength at which they are used. The probe lasers used for diagnostics need to be able to resolve ultrafast transient behavior, and be broadly tunable around 1.55 μm.

Fiber lasers have recently emerged as new sources in this spectral region. Ultrashort pulses can be obtained from these lasers by a passive modelocking technique known as stretched-pulse additive pulse modelocking (SP-APM). We have demonstrated that when pumped with a high power master oscillator power amplifier (MOPA) diode, this SP-APM laser can produce pulse energies up to 2.25 nJ in highly-chirped 1.5 ps pulses, which may then be compressed to less than 90 fs duration and more than 1.5 nJ per pulse[14]. In this mode of operation we have also found that the laser exhibits extremely low amplitude noise (less than 0.1 percent from 0 to 200 KHz), making this an excellent source for pump-probe measurements and also a seeding source for an optical amplifier.

Although these pulse energies are suitable for a variety of experiments, for many applications higher pulse energies are required. In collaboration with W. Gellerman of the University of Utah, we have built an optical amplifier for the purpose of reaching these energies[15]. We have achieved a gain of $> 10^4$ yielding ~ 10 μJ pulses using this system. Our amplifier uses a 2-cm-long piece of KCl:Tl^+ crystal as the gain medium, which is pumped by a Q-switched Nd:YAG at a rate of 1 KHz. To match the pump repetition rate, we have set up a synchronized electro-optic pulse selection system which selects single pulses at the 1 KHz rate from the 40 MHz output pulse train of the SP-APM fiber laser. The amplified pulse passes through the amplifier only twice resulting in excellent beam quality. The amplified pulse has been compressed to 250 fs. With further optimization, we expect to achieve even more energy per pulse and shorter pulses. These energies will allow continuum generation and make possible new time resolved spectroscopy experiments. These experiments will yield new information on the basic physics of optical devices.

Ultrashort Pulse Solid State Lasers

Compact pulse sources

Compact ultrashort pulse sources are of fundamental importance for advances in signal processing, high speed communications, and the investigation of ultrafast nonlinear processes in semiconductor devices and materials. Generally, these sources must be technologically simple, robust and cost effective. While solid state gain media are well suited to meet these criteria, their relatively low gain cross-sections have required the use of fast saturable absorption for modelocking. During the last few years significant advances have been made in the development of fast saturable absorbers utilizing the electronic Kerr effect. Kerr lens modelocking (KLM)[16], in fact, has allowed the generation of the shortest pulses ever produced directly from a laser oscillator[17]. Our group has made several important contributions to the theoretical understanding of these advances[18]. We have further applied our theoretical model of KLM to facilitate optimization of modelocking performance and to allow the extension of this simple pulse forming mechanism to novel resonator geometries. Two Ti:Al_2O_3 KLM lasers in our laboratory, currently producing pulses of $\sim$ 10 fs duration, are direct results of the accuracy of our modeling.

Chirp-free ultrashort pulses are produced in KLM as a result of a self-focusing nonlinearity in the presence of soliton-like pulse shaping arising from self-phase modulation and net negative intracavity group-velocity dispersion (GVD). Negative GVD is most commonly achieved in KLM lasers by use of an intracavity prism pair; however, this places constraints on laser geometry and size. Most KLM lasers have used a folded X- or Z-cavity geometry with the prism pair in one arm of the laser, and repetition rates have not exceeded the 100 MHz range. To achieve higher rep-rates, we have developed a novel and compact dispersion-compensation technique and laser geometry for KLM in solid-state lasers[19]. This design provides built-in and easily adjustable GVD as a direct consequence of the resonator geometry, eliminating the need for prism pairs. We have produced 111 fs pulse durations at a repetition rate of 1 GHz and 54 fs pulses at 385 MHz. The demonstrated geometry has important implications for compact, all-solid-state, femtosecond laser technology, especially because it can readily be extended to the modelocking of diode-pumped lasers.

Cavity-dumped Lasers

Femtosecond laser systems have been widely applied for the study of ultrafast phenomena in physics, chemistry, and biology. An optimum laser source for these studies should have a short pulse duration, wavelength tunability, sufficient pulse energy to permit the investigation of nonlinear effects and sufficient repetition rate to permit the use of averaging techniques for high sensitivity detection. Finally, the laser system should be simple, cost effective, and robust.

Several amplification techniques have recently been developed to extend the range of available pulse energies from Ti:A_2O_3 sources. Regenerative as well as multipass amplifiers

with Nd:YAG and Nd:YLF pump lasers have been demonstrated up to a few kilohertz repetition rates[20]. More recently, argon laser pumped $Ti:Al_2O_3$ amplifiers have produced microjoule energies at repetition rates as high as 450 kHz[21]. However, the requirement of multiple stages and/or multiple pump lasers makes these oscillator-plus amplifier systems complex and relatively expensive. Lastly, the repetition rate of many of these sources is low enough to limit detection sensitivity for ultrafast measurements.

A superior alternative to these laser systems, demonstrated by our group, is cavity dumping of a KLM $Ti:Al_2O_3$ oscillator[22]. This technique allows the generation of 50 fs pulses with pulse energies as high as 100 nJ at variable repetition rates as high as 0.95 MHz. The limitation on the energy currently available from this source arises because of multiple pulse instabilities that occur at high pulse energies as a result of saturation of the KLM saturable absorber action. Decreasing the output coupling of this laser increases the cavity Q but does not enhance the dumped energies since it reduces the pump level at which multiple pulse instabilities occur. We are currently testing techniques such as defocusing the beam within the gain crystal and implementing other dumping mechanisms that do not require a focused beam so that the KLM nonlinearity can be decreased and the instability threshold, thereby, increased.

Flashlamp pumped modelocked $Ti:Al_2O_3$ laser

Nearly all modelocked $Ti:Al_2O_3$ lasers to date have been cw modelocked systems with relatively low pulse energies. Short pulse energies in the range of microjoules to millijoules can be generated using regenerative and multipass amplifier systems. While these techniques achieve excellent performance, they are relatively costly and complex. To achieve higher powers with a simpler system, we have demonstrated a high performance, hybrid-modelocked, flashlamp-pumped $Ti:Al_2O_3$ laser by combining active acousto-optic modulation and fast saturable absorber action from a low temperature multiple quantum well[23]. This laser has produced pulses as short as 4.5 ps with peak powers exceeding 4 MW at a repetition rate of 10 Hz. The flashlamp-pumped $Ti:Al_2O_3$ laser is a simple, economical, high peak power laser source which can be an attractive alternative technology to conventional oscillator-amplifier approaches.

Ultrafast Nonlinearities in Active Semiconductors

We have carried out extensive investigations of nonlinear optics and ultrafast dynamics in semiconductor optical amplifiers (SOAs), observing a number of potentially important phenomena for the first time. The active nature of these devices makes it possible to bias them electronically to produce optical gain or eliminate unwanted long-lived nonlinear components associated with carrier density dynamics. The injected carriers also introduce novel optical nonlinearities that affect ultrashort-pulse propagation in amplifiers and may potentially be used for controlling one optical pulse with another.

Index dynamics in AlGaAs amplifiers

Measurements of gain and index of refraction dynamics have been performed by femtosecond pump-probe over a wide range of photon energy in AlGaAs SOAs[24]. Our broadly-tunable femtosecond Ti:sapphire laser makes it possible to study these nonlinearities throughout the gain region and below band, as well as near the transparency point that has been investigated in detail before[25]. Using a time-division interferometer (TDI), we have performed pump-probe of both the optically-induced transmission dynamics and the index of refraction dynamics. Below band, the transmission dynamics are dominated by the instantaneously responding two-photon absorption that is essentially independent of wavelength and bias current. The index dynamics, however, exhibit a fast, negative dynamic with a bandgap resonant component that varies by more than a factor of ten with bias current and wavelength, in agreement with recent theory[26]. There is also a slower dynamic that provides proof of nonequilibrium carrier heating by free carrier absorption. It is characterized by a 100 fs risetime related to Fermi equilibration and a 1.1 psec relaxation determined by the carrier-lattice energy exchange rate in this material. In above-band measurements also, we have observed large variations of gain and index nonlinearity with bias current. Stimulated emission contributes to either carrier heating or cooling depending upon wavelength, and wavelength-varying gain couples ultrafast index changes to transmission dynamics. The results of all these studies can now be used to improve modeling of ultrafast device characteristics. The strength of the observed nonlinearities, and their dependence on controllable device parameters, make them interesting also for all-optical pulse control and switching applications.

Carrier dynamics in InGaAs strained layer diodes

A new multiple-wavelength femtosecond pump-probe technique developed in our group[27] has made it possible to study the actual, nonequilibrium spectral dynamics in diode amplifiers for the first time[28]. A modelocked Ti:Al_2O_3 laser output is coupled into an optical fiber for self-phase modulation spectral broadening. The light coming out of the fiber is then split into the pump and probe beams. Two separate spectral windowing assemblies allow for independent frequency, pulsewidth, and polarization selection of both pump and probe pulses. The devices investigated were InGaAs/AlGaAs graded-index separate confinement heterostructure SQW ridge waveguide diode lasers, fabricated in collaboration with colleagues at the MIT Lincoln Laboratory. These strained-layer quantum well devices are known for their high power, high efficiency, long lifetime, and low threshold current density.

We were able to map the femtosecond gain dynamics at a range of wavelengths for a fixed excitation pulse. The transient measurements showed a pump-induced transmission decrease in both the gain and loss regions. Around zero time delay, a sharp transmission decrease was observed with a spectral peak around the pump wavelength and a time-resolution-limited recovery. This transient may be attributed to a combination of two-photon absorption, spectral hole burning, and coherent artifacts. Shortly after the pump

pulse, a thermalized carrier distribution with higher temperature and lower concentration is established. Gain depletion throughout the investigated spectral region was observed. For time delays longer than 1 ps, the gain partially recovers as the temperature reaches equilibrium with the lattice. The residual gain changes are produced by the decrease in the carrier population and recover on a much longer time scale.

Under conditions of high carrier concentrations, with the pump wavelength close to the bottom of the gain region, strong spectral hole burning effects were observed. Moreover, an additional blue shift of the spectral hole with respect to the pump wavelength was observed[28]. At lower carrier concentrations with the pump close to the transparency point, spectral hole burning effects were much weaker without any spectral shifts. This new information is important to understanding gain dynamics and its implication on high speed optical device performance such as cross-talk effects, short pulse amplification, and high speed modulation behavior.

Femtosecond dynamic anisotropy

In collaboration with J. Wiesenfield, M.A. Newkirk, U. Koren, and R.M. Jopson of AT&T Bell Labs, we have begun a new set of studies on 1.55 μm SOAs fabricated to have polarization insensitive gain. This polarization insensitivity is achieved by alternating compressive and tensile strained quantum wells in the active region.

Because of polarization selection rules, the compressive wells provide gain only for light polarized in the plane of the wells (TE), while tensile wells provide gain mainly to light polarized perpendicular (TM). The resulting small signal gain is made polarization independent, but the nonlinear dynamics are not necessarily so. By using the single wavelength heterodyne pump-probe scheme developed in our lab[29,30], we are able to investigate these dynamics with any combination of TE or TM pump and probe pulses. Varying the relative polarization between pump and probe changes the measurement sensitivity to processes occurring in different wells. For example, when the pump is TM polarized and the probe TE, the pump will induce interband transitions primarily in the tensile wells and the probe will monitor population in the compressive wells. Thus, we may investigate interwell transport as well as intra-well relaxation. Preliminary experiments have revealed some subpicosecond anisotropy in the dynamic gain response[31]. Future work will quantify these results and provide information about their dependence on quantum-well dimension and other device parameters.

Nonlinear Interactions of CW and Pulsed Light in Optical Fibers

Because of their short duration, modelocked pulses have a broad spectrum. An ultrashort pulse system can be useful in nonlinear spectroscopy for this feature alone without considering its time resolving capability.

We have used the wide spectrum available from self-phase modulated 100 fs pulses from a Titanium Sapphire laser to accurately and directly measure the Raman gain spectrum

of glass optical fibers. The pulses were counter-propagated through 700 meters of fiber against light from a continuous laser whose frequency lay in the middle of the pulses' spectrum. The CW laser modulated the different wavelength components of the pulse train via stimulated Raman scattering. By measuring this modulation spectrum with a spectrometer at the fiber output, we were able to extract the full Raman gain spectrum from 13 THz down to approximately 100 GHz. This approach is much simpler than measuring the gain spectrum point by point with a CW tunable probe laser.

Of course, modelocked pulses are not completely equivalent to a bright broad band incoherent source. The definite relationship between the time and frequency characteristics of the pulses permits observation of new effects which would not occur in nonlinear gain measurements of this type using incoherent light[32]. An effect of this type was discovered in our measurements of Raman gain spectrum at small detuning for several lengths of fiber. At positive Stokes detuning the Raman gain decreases towards zero detuning where it becomes masked by scattered pump light and the Brillouin gain peak at 20 GHz. At negative detuning the mirror image anti-Stokes Raman spectrum is seen with an oscillatory signal superimposed.

These oscillations are a direct result of the connected time and frequency structure of the strongly chirped probe pulses. The chirp on the pulses due to the positive group velocity dispersion (GVD) of glass at 800 nm means that the red frequency components of the pulse travel ahead of the blue components. This implies that at a fixed point in the fiber, the Stokes gain of the probe occurs before the anti-Stokes gain. An excitation left in the glass by the Stokes frequencies can potentially effect the propagation of the anti-Stokes frequencies. For the highly damped Raman modes, no excitation is left behind. However, the central portion of the probe at the lowest frequencies can beat with the pump field to drive sound waves in the glass which result in Brillouin scattering of the backward traveling pump into the direction of the probe. This extra field impresses a time-dependent phase shift on the co-propagating anti-Stokes side of the probe through interference terms in the Kerr effect. The chirp causes this oscillatory phase modulation waveform in the time domain to appear directly in the spectral domain because different frequency components travel with different delays behind the center frequency.

A thermal white light source probe would not reveal this coupling mechanism in its spectrum. Although this phenomena was a nuisance for our Raman experiment, use of such a chirped probe was fortuitous for detecting the effect itself, which may appear more subtly in other situations. For instance, a train of solitons propagating against a CW background will shed these Brillouin wake fields the same as in our experiment. The consequence in this case will be merely a phase or frequency shift on the trailing pulses' spectrum rather than pronounced oscillations. This may contribute excess timing jitter in communications applications or may be judiciously used to cancel soliton self-frequency shift. In analog CW communication systems operating at power levels near the Brillouin threshold, these phase modulations could result in excess noise at channel frequencies detuned much greater than the Brillouin linewidth from the carrier. While these specific ideas are being pursued with further experiments, a more important result of this study is

that small amounts of Brillouin scattering can cause observable indirect effects in certain short-pulse, broad-bandwidth applications where its direct effects can be safely neglected.

Wavelength Shifting by Four-Wave Mixing in Passive Waveguides

To investigate devices for wavelength-division-multiplexed (WDM) optical network applications, we are performing all-optical, wavelength-shifting experiments in semiconductor waveguides at 1.55 μm, in collaboration with H.Q. Le, J.P. Donnelly, S.H. Groves and Eric A. Swanson of Lincoln Laboratory. The wavelength-shifting is accomplished by non-degenerate four-wave mixing (FWM) which uses optical beams at two different input wavelengths (signal-in and pump) to generate output at a third wavelength (signal-out). Since the mixing process depends upon instantaneous field products, there is no response-time limit in the time domain. Instead, the nonlinearity response time of the medium manifests itself as variation of conversion efficiency with magnitude of the wavelength shift. To investigate the possibility of obtaining a relatively flat conversion efficiency over a wide detuning range, we are studying InGaAsP/InP passive waveguides. We have performed both cw experiments and picosecond pulse experiments at 1.55 μm wavelengths, with tunable, synchronized F-center lasers, to evaluate: variations of efficiency with pump wavelength (below band), the effects of phase matching on shifts as large as 130 nm (17 THz), and the limitations on conversion imposed by two-photon absorption. Our data allows us to extract quantitative values for waveguide loss, group-velocity dispersion, and both the real and imaginary parts of $\chi^{(3)}$. Two-photon absorption, strictly a liability in nonlinear index switching, can actually enhance FWM conversion efficiency to some extent. We have analyzed this theoretically and quantified it experimentally[33]. In a 7.5-mm-long passive InGaAsP single quantum well waveguide, we have achieved a conversion efficiency for picosecond pulses of -11 dB for a wavelength shift of 20 nm. In a waveguide with an effective mode area of 3×10^{-3} cm^2, this requires a pump pulse energy of 10 nJ in 10 ps. The waveguide structures are fabricated at MIT Lincoln Laboratory. Future plans call for optimizing design of the waveguide shifters using multiple quantum wells and incorporating a p-n junction for carrier sweep-out.

Nonequilibrium Electron Dynamics in Metals

Studies of interactions between free carriers and their environment constitute one of the major problems of solid state physics. This has been addressed directly in the time domain employing femtosecond techniques, both in semiconductors and metals. Of particular interest in the case of metals is the display of very high electron density whose behavior can be modeled on a relatively simple basis. In previous experiments[34], it was demonstrated that it is possible to create and probe transient nonequilibrium electron populations using ultrashort laser pulses. In these experiments, it was assumed that electron-electron interactions were sufficiently fast to thermalize the electron gas on the order of, or shorter,

than the pulse pump duration. This assumption was made even though some deviations from the instantaneous response were observed[35]. More recent investigations using transient photoemission have demonstrated the existence of non-Fermi electron distribution with thermalization times as long as 600 fs[36]. These results were observed in gold foil for large changes of the electron temperature (of the order of 400°K) with a limited time resolution. Similar conclusions were drawn at lower laser fluence by analyzing the temperature dependence of the optically measured electron-photon interaction time in gold and silver[37].

Femtoscond studies of electron thermalization in metals were conducted using transient thermomodulation transmissivity and reflectivity. Studies were performed using a tunable multiple-wavelength femtosecond pump-probe technique in optically thin gold films in the low perturbation limit. An infra-red (IR) pump beam was used to heat the electron distribution and changes in electron distribution and electron temperature were measured with a visible probe beam at the *d*-band to Fermi-surface transition[38]. These studies show that the subpicosecond optical response of gold is dominated by delayed thermalization of the electron gas. This effect is particularly important far off the spectral peak of the reflectivity or transmissivity changes, thus permitting a direct and sensitive access to the internal thermalization of the electron gas.

Measurements using a high stability, high repetition rate, and tunable, modelocked $Ti{:}Al_2O_3$ laser, show evidence for non-Fermi electron distribution. This distribution was measured to have an electron thermalization time of the order of 500 fs and an electron-lattice cooling time of 1 ps, independent of laser fluence in the range of 2.5 mJ/cm^2. At energies close to the Fermi surface, longer thermalization times $\sim 1 - 2$ ps were observed. These results are in agreement with a more sophisticated model based on calculations of the electron-thermalization dynamics by numerical solutions of the Boltzman equations[39]. This model quantitatively describes the measured transient optical response during the full thermalization time of the electron gas to be of the order of 1.5 ps. Finally, the theoretical and experimental techniques developed in this study can form the basis for investigating and understanding nonthermal electronic effects in a wide range of materials.

Time-Gated Scanning Tunneling Microscopy

Since the invention of the scanning tunneling microscope (STM) in 1982, scanning tunneling microscopy has become a well established technique that allows us to study surfaces with very high spatial resolution down to atomic dimensions. However, the temporal resolution of a conventional STM is limited to microseconds. Today, femtosecond laser technology enables measurement of the highest time resolution possible of solid state phenomena. However, the spatial resolution is limited to microns by a laser spot size. A combination of STM with ultrafast time resolution of femtosecond lasers would enable us to develop an instrument capable of performing surface measurements highly localized in both spatial and temporal domain. Recently, a number of approaches to this problem have been taken[40].

In an STM, a sharp metal tip is positioned a few angstroms away from the surface under investigation. An electrical bias is applied between the tip and the surface. When the tip is close to the surface, a small tunneling current will flow from the tip to the surface. The size of this current depends exponentially on the distance between the tip and the sample. If we heat an electron distribution with a laser pulse, we would expect the tunneling of heated carriers to be different from that of a cold equilibrium distribution.

In our experiment, a Pt-Ir tip is positioned above an Au surface. Amplified femtosecond laser pulses are applied to the tip-sample junction of an STM and the effect of the laser pulses on the tunneling current is monitored. Several effects contribute to the laser induced tunneling current. They include three-photon photoionization and carrier heating, which in turn leads to thermally assisted photoionization and thermally assisted tunneling[41]. Time resolution is achieved by splitting a laser pulse into two pulses of equal energy and varying time delay between them. As a result, a femtosecond interferometric cross correlation is obtained. We have investigated the dependence of different components of the laser assisted tunneling current on a number of parameters including laser intensity and polarization and the STM tip-sample bias voltage.

Coherent Phonons in Solid-State Materials

Pulses of laser light, which are short compared to the optical phonon period, can be used to impulsively generate the coherent lattice vibrations in solid state materials. Our current interest in coherent phonons is two-fold: (1) use time-resolved femtosecond spectroscopy to further our understanding of lattice and electron dynamics, and (2) exploit the coherent phonon phenomenon to modulate the physical characteristics of the material at THz frequencies. For semimetals and narrow-gap semiconductors, the reflectivity modulations due to coherent ion motion have been particularly large (on the order of 10 percent in some materials) and have suggested significant modulation of the material's electronic structure. In these cases, we have determined that the coherent phonon excitation is caused by a mechanism we call Displacive Excitation of Coherent Phonons (DECP)[42,43].

The DECP mechanism uses the pump pulse to promote electrons in the solid from bonding orbitals to anti-bonding orbitals, thereby changing the ionic potential's coordinate of minimum energy. Because the lattice cannot respond adiabatically to this rapid displacement of the ionic equilibrium coordinate, the lattice is set into oscillation about the new equilibrium. With the DECP mechanism, the generated coherent phonon can be large enough to modulate significantly the energy bands of a solid through a deformation coupling[44]. Energy band modulation is manifested through transient reflectivity modulations as large as 12 percent in semiconducting Ti_2O_3 at the A_{1g} optical phonon frequency ($\sim$ 7 THz). In experiments involving high excitation intensities, it is observed that the optical phonon is screened by the photoexcited carriers, resulting in an initial down-shift of the optical phonon frequency relative to the value found through spontaneous Raman scattering.

In most femtosecond studies of carrier dynamics, the induced optical changes are the

result of carrier distributional changes that evolve with energy and momentum loss to the lattice. In the present experiment, the induced optical changes are dominated by the lattice and we measure changes in the phonon frequency as affected by the ions' interaction with the excited carriers. In the most direct way possible, we observe electron-phonon interactions as experienced from the point of view of the lattice itself.

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